Pestic. Sci. 1998, **54**, 99–112

A Bayesian Approach to the Analysis of Environmental Fate and Behaviour Data for Pesticide Registration

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(Received 19 November 1997; accepted 4 June 1998)

Abstract: With the harmonisation of data requirements for pesticide registration under EC Directive 91/414 there is need for progress on the techniques used to analyse such data and so help make consistent the judgements applied by national regulatory authorities. This paper proposes a Bayesian technique for combining data from environmental fate and behaviour studies of pesticides in soil. The method uses expert knowledge, based on degradation and adsorption data, and logistic regression methods to form a prior probability distribution for the probability that a given compound leaches. Results from lysimeter experiments are used update the prior knowledge. Data for the compounds bentazone and triclopyr are used to illustrate the techniques. The advantages of the methodology and its implications for the pesticide registration procedure are discussed in the light of possible advances using modern Bayesian statistical techniques and mathematical models. © 1998 Society of Chemical Industry

Pestic. Sci., **54**, 99–112 (1998)

Key words: Bayesian statistics; classification; expert knowledge; groundwater; leaching; logistic regression; lysimeters; pesticide contamination

1 INTRODUCTION

The publication of EC Directive 91/414 began the process of harmonisation of pesticide registration throughout the countries of the European Union (EU), and this work has been extended beyond Europe through the work of the OECD.[1] Included in Annex II of this Directive is the list of common data requirements, covering such topics as toxicology and environmental fate and behaviour, to be applied when considering a plant-protection compound for registration or re-registration. Under the Directive, member states of the EU retain their national regulatory authorities, and they are to move to full implementation of the Directive within ten years of its publication. Even with the development of 'uniform principles' the assessment of whether or not a new compound represents an unacceptable risk remains a matter of expert judgement (UK Pesticide Safety Directorate, pers. comm.).

With regard to the environmental fate and behaviour of pesticides in soils, Directive 91/414 requires: the measurement of adsorption and desorption of the compound; the rate and route of degradation; and a measure of the compound's mobility, all of which must be measured in at least three soils. In addition the extent and nature of any bound residues must also be measured. Standard techniques have been or are being developed for the measurement of these, for example for the mobility of a compound by use of lysimeters.[2]

When working with a heterogeneous medium such as soil, it would be difficult and impractical to produce a set of measurements that could adequately cover the diverse range of soil properties and situations that may be encountered when applying pesticides. Computer simulation models have, therefore, been seen as a means of extending the range of possible scenarios for which

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 Pestic. Sci. 0031–613X/98/$17.50. Printed in Great Britain

environmental fate and behaviour information is available. The UK Pesticide Safety Directorate (PSD) allows evidence from simulations to be included in a registration submission provided that sufficient data accompany the submission to the authority to allow for its validation. This authority prefers the use of tried and tested models (e.g. LEACHM[3]) rather than state-of-the-art models that may include such physical processes as macropore flow (e.g. MACRO[4]) (Jarvis and Griffin, 1996, unpublished). Laskowski *et al.*[5] have called for greater use of probabilistic modelling of pesticide leaching because soil variability is inherent in these models. Whether or not computer simulation evidence is included, it is still necessary to apply expert knowledge when judging whether or not a compound should be registered for use. The purpose of this paper is twofold: first, to outline a simple technique for combining and analysing data from pesticide registration studies as a means of better understanding the probability that a given compound will present an unacceptable risk; and secondly, to discuss the limitations of present practice for handling environmental fate and behavior data as part of registration studies.

2 BAYESIAN METHODOLOGY

Bayesian statistics provides the only formal coherent framework for quantifying expert knowledge and combining it with data.[6–11] In this approach, expert judgements are summarised in the form of probability distributions over the quantities of interest. These summaries represent probabilistically the initial, or *prior* state of knowledge. After experimental data have been collected, the prior state of knowledge may be formally updated using Bayes' theorem to combine the prior knowledge with the data. The result is a *posterior* state of knowledge, summarised in the form of updated probability distributions over the quantities of interest. In this paper, the quantity of interest we focus upon is the probability that a given pesticide will leach when applied in a soil environment.

2.1 Relevant data

We wish to reduce our uncertainty about the probability θ that a particular pesticide will leach into groundwater, using data from lysimeter experiments. We assume that the relevant data from the results of each lysimeter experiment will be whether or not the pesticide is observed to leach, relative to a specified threshold. In a series of n such independent experiments, denote by r the number of experiments in which the pesticide is observed to leach, and by $s = n - r$ the number of experiments where leaching is not observed. Before the experiments are run we can only say that r can range from 0 to n. The probability of obtaining r leachers and s non-leachers from n lysimeter experiments is given by the *binomial* distribution:

$$p(r \mid n, \theta) = \frac{n!}{r!s!}\,\theta^r(1-\theta)^s \quad r = 0, 1, \ldots, n \qquad (1)$$

where the notation $p(r \mid n, \theta)$ is used to show that, to calculate the probability, we must know not only the value of n but also that of θ. Of course, if n and θ are known we can calculate the probability of obtaining any specific value of r. However, in this problem we have a situation in which n and r are known and it is the unknown θ which is the quantity of interest. To study this, we consider (1) as a function of θ, when it is known as the *likelihood* function, and provides the link between θ and the data. In general, the likelihood may be regarded as the information about θ provided by the data alone. One value θ_1 is better supported by the data than another value θ_2 if the likelihood of θ_1 is than that of θ_2.

2.2 Prior knowledge

Although the exact value of θ is unknown, there may be prior information concerning its value, in the form of expert knowledge. Such information may stem, for example, from general knowledge that the pesticide may be similar in action to another well-studied pesticide, or from previous experimentation. Bayesian statistics uses a probability distribution for θ—called a *prior distribution*—to quantify such knowledge prior to relevant data collection. We will model prior knowledge using a *beta distribution*, which is specified by a probability density function of the form

$$p(\theta \mid a, b) = \frac{1}{B(a, b)}\,\theta^{a-1}(1-\theta)^{b-1} \quad 0 \leqslant \theta \leqslant 1 \qquad (2)$$

where the values of a and b are chosen in such a way that this distribution describes current knowledge about θ at the stage prior to obtaining relevant data r, n; and $B(a, b)$ is a constant (not depending on θ) which ensures that the area under this curve is 1.

The beta distribution is particularly appropriate for this situation, for a number of reasons. It is very flexible and can model accurately a wide range of different forms of prior knowledge about θ. Further, it is restricted to values between 0 and 1 for θ, as is appropriate for a variable representing a probability. From a mathematical perspective, the posterior distribution obtained when this prior distribution is combined with data from a binomial distribution is also a beta distribution, but with different values of a and b. This is a very desirable property in that we then have a new beta

distribution to form the prior knowledge for any further data collection.

The prior mean and standard deviation of θ are

$$\frac{a}{a+b} \quad \text{and} \quad \sqrt{\frac{ab}{(a+b)^2(a+b+1)}} \tag{3}$$

For example, choosing $a = 3$, $b = 2$ leads to a prior distribution with mean 0·6 and standard deviation 0·2, reflecting prior knowledge concerning the value of θ, and uncertainty about it (Fig. 1).

In general, the process of choosing a prior distribution to describe prior knowledge (before additional information becomes available) is called *elicitation*. If a beta distribution is appropriate, we have seen above that the elicitation process simplifies to choosing values for a and b. There is a rich literature on this important topic in Bayesian statistics.[12] Further discussion on elicitation is addressed below.

2.3 Combining prior knowledge with data

Once a prior distribution has been specified, it can be updated using observed data. In this setting, the data are the numbers r out of n lysimeter experiments which result in the pesticide leaching.

Combining the prior knowledge, in the form of a beta distribution with parameters a and b, with data as specified above, leads to a posterior distribution which is also a beta distribution, but with parameters $a + r$ and $b + s$. The posterior mean and standard deviation of θ are

$$\frac{a+r}{a+b+n} \quad \text{and} \quad \sqrt{\frac{(a+r)(b+s)}{(a+b+n)^2(a+b+n+1)}} \tag{4}$$

The posterior mean is a new estimate of θ. For example, with $a = 3$, $b = 2$, $n = 4$, $r = 3$, $s = 1$ we obtain for θ a posterior mean of 0·67 and a posterior standard deviation of 0·15. For this hypothetical example, the result of combining prior knowledge with experimental data is to increase slightly from 0·60 to 0·67 the probability that the compound leaches, with a corresponding increase in precision as the standard deviation falls from 0·20 to 0·15. As well as an estimate of θ, knowledge is often summarised by reporting an interval to which θ belongs with a stated probability. Such an interval, sometimes called a *Bayesian confidence interval*, can be determined numerically or found from tables.[13]

2.4 Assessing the value of prior information

The updating process described above gives a plausible interpretation of a and b in the prior distribution for θ. It can be seen that a plays a role similar to that of r, the observed number of experiments which result in leaching and, likewise, b plays a role similar to that of s, the observed number of experiments in which the pesticide does not leach. This allows interpretation of a and b in terms of imaginary data, in the sense that the prior distribution models knowledge that may be regarded as equivalent to $a + b$ imaginary lysimeter experiments in

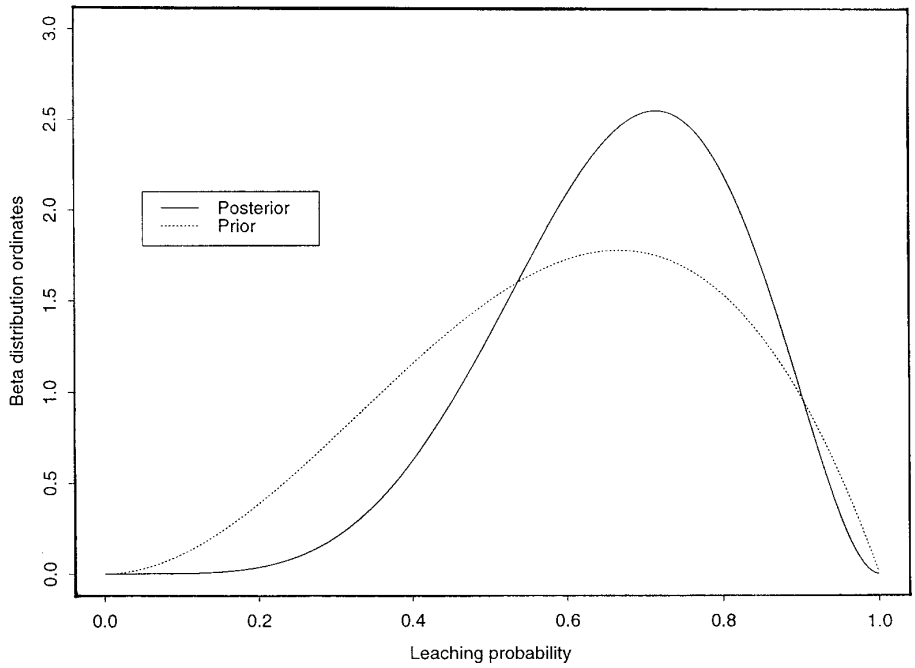

Fig. 1. The beta prior probability density function $p(\theta \mid a = 3,\ b = 2)$, with prior mean 0·60, and the beta posterior probability density function $p(\theta \mid a + r = 6,\ b + s = 3)$, with posterior mean 0·67. The evidence from the data, obtaining leachers from three of four lysimeter experiments, gives a posterior probability distribution which is somewhat to the right of the prior probability distribution, and with a smaller standard deviation.

which a result in leaching and the remaining b do not. Of course, in general, a and b do not have to be integers. This interpretation of a and b is particularly useful for elicitation.

Bayesian methodology combines information from two sources: prior knowledge and actual data. As $a + b$ plays a role similar to that of n, we can effectively determine the relative weight of information from each of these two sources. In the example above, the value of the prior information amounts to $a + b = 3 + 2 = 5$ lysimeter experiments. The data then provide further evidence from $n = 4$ actual lysimeter experiments, so that the prior knowledge and data respectively contribute 5/9 and 4/9 of the information used to form the combined posterior judgement. Obtaining an appropriate balance here is an important issue we address later.

The analyses of information and related statistical inferences do not stop with the posterior distribution given above. If further data become available, the updating process is analogous to that from prior to posterior above; for example, if the new data comprise a further n' lysimeter experiments (on the same compound under the same conditions) of which r' result in leaching and the remaining s' do not, then the $a + r$ and the $b + s$ in the posterior distribution above are simply replaced by $a + r + r'$ and $b + s + s'$; and so forth, when further such data become available. The posterior distribution for θ after two updating stages is identical to that had we been given all the data, $r + r'$ and $s + s'$, at the same time.

Prediction is readily catered for within the Bayesian framework. For a particular pesticide we may be interested in assessing the probability that a future lysimeter experiment will result in it leaching, given that it leached in r out of n previous lysimeter experiments. In this particular simple situation, this predictive probability turns out to be the mean of the posterior distribution of θ, given that the pesticide leached in r out of n previous experiments. In other more complicated settings, the predictive probability distribution may not be so intuitively simple but in principle it can always be computed.

3 APPLICATION TO PESTICIDE CLASSIFICATION

3.1 Introduction

We now apply the methodology described in the previous section to the problem of using data generated as part of the pesticide registration process. To apply the methodology, we need a way of quantifying our prior knowledge about the environmental fate of pesticides. Various workers have suggested simple pre-screening or classification methods for the environmental fate of pesticides based on laboratory-derived parameters (especially degradation rate and the adsorption coefficient or related parameters). Gustafson[14] proposed an index based on soil degradation half-life and adsorption coefficient (K_{oc})[15] as a means of distinguishing between pesticides that would and would not pollute groundwater. The threshold values were based on observations of which compounds were found in US groundwater, and the work is an extension of earlier work by the California Department of Food and Agriculture.[16] Similar methods have been proposed by Cohen *et al.*[17] and Jury *et al.*[18] Helling and Turner[19] and McCall *et al.*[20] have both proposed classifications of soil mobility based solely on a compound's K_{oc} value. The Soil Survey and Land Research Centre (SSLRC) has suggested a classification of a compound's mobility based on the K_{oc} value and a parallel classification of persistence of the compound based on the soil half-life.[21] Nicholls[22] has combined several of these methods into an expert system for pre-registration screening. All these provide a method of forming prior evidence based only on data derived from laboratory experiments.

We propose an alternative means of classification, based on a statistical technique known as *logistic regression*, which is widely accepted as being the appropriate way of handling binary response data.[23] This technique, first introduced by Berkson,[24] depends on transforming the (0,1) interval to $(-\infty,\infty)$ for the purpose of estimating a relationship, and then transforming back to (0,1) so that the end result of the prediction equation is a *probability* for a binary outcome. Thus, logistic regression is characterised by relating one or more explanatory variables (for example adsorption coefficient K_{oc} and pesticide soil half-life $t_{1/2}^{soil}$) to a binary response variable (for example a pesticide may be observed to leach or not). The resulting logistic regression equation can then be used to estimate the probability that a particular pesticide leaches. Collett[23] describes the algorithm to perform logistic regression and discusses the merits of various statistics packages for carrying out logistic regression. We used the package S-Plus (for example, Venables and Ripley[25]) to perform the analysis, but many other commonly available statistics packages, for example GLIM[26] and SPSS,[27] also perform logistic regression.

3.2 Using logistic regression to predict pesticide leachability

A classification of 29 pesticides into leachers, non-leachers, or transitional, together with an adsorption coefficient K_{oc} and soil half-life $t_{1/2}^{soil}$ in days for each pesticide, was reported by the California Department of Food and Agriculture[16] (CDFA). (Table 1). The CDFA report contained incomplete information on 15 other pesticides (eight were classified as leachers and seven as non-leachers), which we have omitted from our analysis.

TABLE 1
Adsorption Coefficient K_{oc} and Soil Half-Life $t_{1/2}^{soil}$ in Days for 22 of 44 Pesticides reported by the California Department of Food and Agriculture (CDFA), and whether or not the CDFA classified the Pesticides as Leachers[a]

Pesticide	*Leacher*	*Adsorption rate* K_{oc}	*Soil half-life* $t_{1/2}^{soil}$	*Estimated leaching probability*, θ
Aldicarb	Yes	17	7	0·9835
Atrazine	Yes	107	74	0·9854
Diuron	Yes	389	188	0·9035
Metolachlor	Yes	99	44	0·9529
Oxamyl	Yes	26	8	0·9500
Picloram	Yes	26	206	0·9999
Prometryn	Yes	614	94	0·2005
Simazine	Yes	138	56	0·9243
Chlordane	No	19 269	37	7·32 e-08
Chlorothalonil	No	1380	68	0·0053
Chlorpyrifos	No	6085	54	1·36 e-05
2,4-D	No	53	7	0·5009
DDT	No	213 600	38 200	0·0051
Dicamba	No	511	25	0·0109
Endosulfan	No	2040	120	0·0066
Endrin	No	11 188	2240	0·0595
Heptochlor	No	13 330	109	6·02 e-06
Lindane	No	1727	569	0·5105
Phorate	No	1660	38	5·29 e-04
Propachlor	No	794	4	1·21 e-05
Toxaphene	No	95 816	9	4·05 e-12
Trifluralin	No	7950	83	1·77 e-05
Alachlor	Transitional	161	14	0·1181
Carbaryl	Transitional	423	19	0·0098
Carbofuran	Transitional	55	37	0·9903
Dieldrin	Transitional	12 100	934	0·0039
Dinoseb	Transitional	5900	30	2·82 e-06
Ethoprop	Transitional	26	63	0·9998
Fonofos	Transitional	5105	25	2·82 e-06

[a] Of the remaining 22 pesticides, seven were classified as transitional (the evidence concerning their potential to contaminate was inconclusive or conflicting), and 15 are not shown as they had no value reported for adsorption coefficient or soil half-life.

The accuracy of the analysis which follows would be improved if full information for these 15 became available.

A logistic regression using two explanatory variables K_{oc} and $t_{1/2}^{soil}$ was carried out for the binary response (leacher, non-leacher). Both explanatory variables were transformed to a log-scale as the explanatory variables cover ranges which differ by orders of magnitude. All logarithms in this paper are to base *e*. Using the given 22 observations, the prediction equation for log $(\theta/1-\theta)$, the logit (or log odds) of the probability θ that a pesticide with given values of K_{oc} and $t_{1/2}^{soil}$ leaches, turns out to be

$$\text{estimated logit}(\theta \mid K_{oc}, t_{1/2}^{soil}) = 8{\cdot}72 - 3{\cdot}59 \log(K_{oc}) + 2{\cdot}86 \log(t_{1/2}^{soil}) \quad (5)$$

The standard errors for the estimated coefficients in (5) are 4·60, 1·96 and 1·78, respectively. Traditional significance tests should be interpreted with care, but here indicate that the overall model fit is good, and that both explanatory variables are useful, with log (K_{oc}) slightly more important than log $(t_{1/2}^{soil})$.

The transformation back to estimated probabilities gives

$$\hat{\theta} = \frac{e^{8{\cdot}72 - 3{\cdot}59 \log(K_{oc}) + 2{\cdot}86 \log(t_{1/2}^{soil})}}{1 + e^{8{\cdot}72 - 3{\cdot}59 \log(K_{oc}) + 2{\cdot}86 \log(t_{1/2}^{soil})}} \quad (6)$$

The estimated values of θ for each of the 22 pesticides used to fit the prediction equation can now be calculated (Table 1). For example, on the basis of adsorption coefficient and soil half-life alone, the pesticide aldicarb (a known leacher) has an estimated probability of about

98% of being a leacher, whilst chlorothalonil (a known non-leacher) has a corresponding estimated probability of only 0·53%.

A crude approach to classification would be to classify a pesticide as a leacher if its estimated probability of leaching exceeds some threshold, such as $\theta > 0{\cdot}5$. For the sake of argument, using a 50% threshold and assuming we take the findings of the CDFA at face value, the compounds 2,4-D and lindane are marginally misclassified as leachers. The known leacher prometryn is also misclassified and has an estimated leaching probability of about 20%, which is quite distant from the crude leaching/non-leaching threshold. For the remaining 19 compounds the estimated probabilities are all close to one or zero, leaving little room for doubt that the compounds are leachers or non-leachers, respectively.

Estimated leaching probabilities for combinations of K_{oc} and $t_{1/2}^{soil}$ are shown *via* isoprobability lines in Fig. 2. Estimated probabilities decrease or increase steeply as we move away from the solid line indicating 50% probability of leaching. It should be noted that the estimated logistic regression coefficients are quite highly correlated, and so there is some uncertainty about the slope of the isoprobability lines. We address this and related issues below. The ability of the logistic regression to classify the pesticides according to probabilistic thresholds is thus apparently an improvement over Gustafson's method[14] in that there is a more substantial and evident separation between leachers and non-leachers,

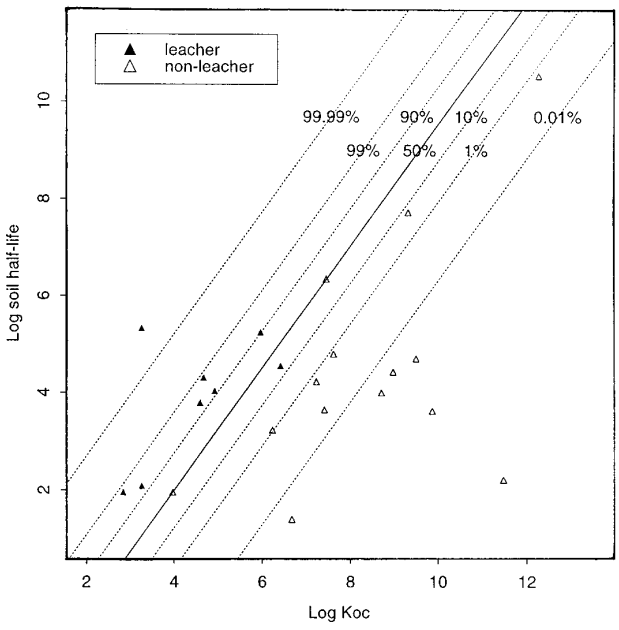

Fig. 2. Crude classification of pesticides by estimated probability of leaching. Pesticides are plotted as leachers or non-leachers together with their corresponding values of log K_{oc} and log $t_{1/2}^{soil}$. Estimated isoprobability lines are also shown. All compounds below (above) the solid line could be classified as non-leachers (leachers) in having probability smaller (greater) than 50%.

together with the advantage that the approach yields a probability rather than a relative score.

Rather than establishing a simple threshold, for regulatory purposes it may be better to set lower and upper thresholds. Actual thresholds would need to be set by regulatory authorities to take into account the utilities of the risks posed by rejecting safe pesticides unnecessarily and of accepting pesticides which do contaminate. For the sake of argument, suppose that we set lower and upper thresholds at 10% and 90% leaching probabilities. Based on the two chemical properties K_{oc} and $t_{1/2}^{soil}$, the model would classify seven of the eight known leachers as leachers and twelve of the fourteen non-leachers as non-leachers. The remaining three compounds have estimated leaching probabilities between 10% and 90%, and could be judged as compounds for which the evidence is insufficiently compelling to classify them either way.

For compounds for which the prediction seems poor, it may be that their leaching behaviour cannot be adequately described by their values of K_{oc} and $t_{1/2}^{soil}$ alone, and that some other mechanism is responsible. There may also be special causes for the compound being detected in groundwater, for example direct hydraulic conduits from the surface. We address model deficiencies in Section 4.

Using the fitted logistic regression model (6), the seven transitional compounds (Table 1) may be classified. There are no obviously marginal cases. Carbofuran and ethoprop both have estimated probabilities $\theta > 0{\cdot}99$ of being leachers. Alachlor has estimated leaching probability of 0·1181, which may be large enough to suggest that we should be careful about classifying it as a non-leacher. The other compounds have very small estimated leaching probabilities and this evidence points to these being non-leachers. For these transitional compounds, the classification based on Gustafson's GUS score is similar, except that the distinction between leachers and non-leachers using the GUS score is less distinct, and that alachlor would remain unclassified.[14]

It is worth remarking that this is a simple way of converting current knowledge into probability form suitable either for the aim of pre-screening compounds for pesticide registration, or for generating prior information in a form suited to the Bayesian approach. Other choices of covariates, or transformations of them, may improve prediction. However, we feel that the proposed method is not only simple, but at least as physically intuitive as other methods. For example, DDT continues to be classified as a non-leacher by Gustafson and the CDFA, and is very far inside their suggested boundaries. The problem with DDT is that it has an extremely long soil half-life, which, coupled with a large K_{oc} implies that it is liable to pollute *via* mechanisms not addressed in this classification, such as sediment run-off.

The proposed method is practicable and realistic, but the results depend on the accuracy of data on only 22 compounds and their presence or absence in Californian groundwater. The reliability of the methodology of the approach (and any other approach based on these data) would be much enhanced by obtaining data on more compounds and under different climatic conditions. Examples of other sources are available.[28,29]

3.3 Assessing the value of the prior information

Suppose that we have been through the process described above, and that we have a new pesticide to consider. We measure its adsorption coefficient K_{oc} and soil half-life $t_{1/2}^{\text{soil}}$, and insert these values into (6) to estimate its leaching probability. The estimate can be used for pre-screening, but we wish to use it for our Bayesian approach, which requires that we generate a prior probability distribution for the leaching probability for each compound, based on its K_{oc} and $t_{1/2}^{\text{soil}}$. This could be done in a number of ways, but we suggest the following method, which depends on making a judgement as to the reliability of the pre-screening. That is, for a given pesticide would we rather have (a) the results of m lysimeter experiments, or (b) the CDFA data and the estimated leaching probability arising from them? We assess the value of (b) as worth at least one and probably more than one lysimeter experiment, as it is based on genuine field observation of environmental fate. On the other hand, there are uncertainties in how reliable the prediction for a new compound will be, and we must take into account that the CDFA observations are for the climatic conditions in limited areas of California rather than for proposed application of the new compound in other soil and climate areas. Certain EU member states currently advise that pesticide registration requires evidence from three lysimeter experiments. It can be argued that the evidence from three regulatory experiments should not be dominated by our prior evidence, and this suggests that the value of our prior evidence should not exceed three lysimeter experiments. These considerations lead us to suggest that the value of our prior evidence is approximately $m = 2$ lysimeter experiments for a new compound, and we use this value for our example later. The choice is not restricted to $m = 2$: in general, m can be any positive value and not necessarily an integer.

For a given new compound we have an estimated leaching probability $\hat{\theta}$ from the logistic regression, and we feel that this information is worth m observations, each observation being a lysimeter experiment. We need now to convert these two pieces of information into a beta prior distribution for the true probability that a compound leaches. From Section 2.4, it follows that we need to choose the beta parameters a and b so that $a + b = m$ and $a/a + b = \hat{\theta}$. These lead to $a = m\hat{\theta}$ and $b = m(1 - \hat{\theta})$. We have suggested $m = 2$ which leads to a beta prior distribution with parameters

$$a = 2\hat{\theta} \quad \text{and} \quad b = 2(1 - \hat{\theta}). \tag{7}$$

For general m, this prior distribution has mean $\hat{\theta}$ and standard deviation $\sqrt{\hat{\theta}(1 - \hat{\theta})/m + 1}$, from which it should be clear that the larger the value of m suggested, the more precise will be our prior distribution. Small values for m, for example $m = 0{\cdot}1$ (thus judging the prior evidence as worth only a fraction of a lysimeter experiment) would result in a very diffuse prior distribution, with the effect that any conclusions drawn will depend almost entirely on the actual lysimeter experiments.

Other choices for m may be appropriate in different contexts; for example, we may be considering an entirely new pesticide with few known properties or, on the other hand, we may be considering a compound presented for re-registration where information is available. In the former case, we want to place more reliance on actual lysimeter experiments, so we choose m to be quite small, whereas in the latter, we should be prepared to value our prior evidence quite highly, and so choose a larger value for m. It should be emphasised that such choices and judgements in a general sense cannot be avoided; it is a great advantage of the Bayesian approach that their role is formalised. In our view this not only leads to better, more reliable, analyses and inferences, but to established formal procedures in which the role of any prior information or expert judgement is made clear, and can be agreed or challenged as those involved see fit.

3.4 Example 1: Assessing the environmental fate of triclopyr

To illustrate the mechanics of the approach, consider the compound triclopyr, for which there are reported values of $K_{\text{oc}} = 41$ and $t_{1/2}^{\text{soil}} = 6$,[30] generated using standard methods.[2]

3.4.1 Generating a prior distribution

First, (5) and (6) are used to obtain an estimated probability of leaching for this compound of 0·619. This is consistent with the corresponding GUS score of 1·86 which would, according to Gustafson, locate this compound in the region in which transition occurs from leachers to non-leachers. Secondly, (7) is used to generate the parameters for the beta prior distribution and so obtain $a = 1{\cdot}24$, $b = 0{\cdot}76$. We can use statistical tables for Bayesian confidence limits for a binomial parameter[13] to find a highest density prior probability interval for the true leaching probability θ, interpolating in the tables where appropriate. We find that a 90% interval for θ in this case is about 0·214–0·882. Therefore, the prior evidence suggests that triclopyr leaches

with estimated probability $\hat{\theta} = 0{\cdot}619$, and also that there is a 90% probability that this probability lies between 0·214 and 0·882.

3.4.2 Updating knowledge

In this case two separate lysimeter experiments were undertaken on triclopyr: the annual leachate concentrations in both lysimeters were below 0·1 μg litre^{-1}. In the notation of Section 2.1, there are thus $n = 2$ observations, $r = 0$ leachers and $s = 2$ non-leachers. We now combine the data with the prior evidence in the manner described in Section 2.3 to arrive at a posterior beta distribution which has parameters $a + r = 1{\cdot}24$, $b + s = 2{\cdot}76$. This leads to revised leaching probability of $(a + r)/(a + b + n) = 0{\cdot}310$, with a corresponding 90% Bayesian confidence interval of 0·043–0·504. The evidence, after combining prior knowledge and data, is now more in favour of triclopyr not being a leacher.

3.5 Example 2: Assessing the environmental fate of bentazone

The herbicide bentazone[31,32] has K_{oc} between 13·3 and 175·6 litre kg^{-1} and soil half-life between three and 21 days;[31] however, it has been suggested that $K_{oc} = 34$ litre kg^{-1} and $t_{1/2}^{soil} = 20$ days.[33] In terms of the SSLRC classification this compound is very-mobile to mobile, but impersistent to slightly persistent. In this example we demonstrate again the mechanics of the approach, but this time broadening the analysis to take into account possible interpretations. Note in particular that the values for K_{oc} and $t_{1/2}^{soil}$ occupy a wide region across the centre of the prediction equations.

3.5.1 Generating a prior distribution

Estimated leaching probabilities using (5), (6) at the corners of the region and at the suggested value are shown in Table 2. GUS scores and the GUS classification are shown for comparison. Next (7) is used to generate the parameters for the beta prior distribution. For example, for $K_{oc} = 34$ and $t_{1/2}^{soil} = 20$ we obtain $a = 1{\cdot}98$, $b = 0{\cdot}02$. From tables[13] we find that a 90% probability interval for θ in this case is about 0·458–0·997. Therefore, assuming that the adsorption coefficient and soil half-life for bentazone are 34 and 20 respectively, the prior evidence suggests that bentazone is a leacher with estimated probability $\hat{\theta} = 0{\cdot}990$, and also that there is a 90% probability that this probability lies between 0·458 and 0·997. Similar prior summaries can be calculated for the corners of the region (Table 2).

3.5.2 Updating knowledge

Four separate lysimeter experiments were undertaken on bentazone: in each, the annual leachate concentrations were below 0·1 μg litre^{-1}.[32] Thus, in the notation of Section 2.1, $n = 4$ observations, $r = 0$ leachers and $s = 4$ non-leachers. Assuming that the adsorption coefficient and soil half-life for bentazone are 34 and 20 respectively, the posterior beta distribution (4) has parameters $a + r = 1{\cdot}98$, $b + s = 4{\cdot}02$. The revised leaching probability is $(a + r)/(a + b + n) = 0{\cdot}333$, with a corresponding 90% Bayesian confidence interval of 0·106–0·628. The evidence, after combining prior knowledge and data, is now more in favour of bentazone not being a leacher. If we repeat the analysis for the corners of the adsorption coefficient and soil half-life region for bentazone, as shown in the final columns of Table 2, we arrive at similar conclusions.

Purely to illustrate how the posterior distribution changes shape to reflect different data outcomes, the possible posterior distributions for the leaching probability of bentazone are drawn assuming that bentazone has adsorption coefficient 34 and soil half-life 20 days, and that there are $n = 4$ lysimeter experiments (Fig. 3). For no lysimeter experiments for which bentazone leached, $r = 0$, the posterior distribution has the bulk of its mass to the left of $\theta = 0{\cdot}5$, so that the data and the prior evidence have combined to indicate a low posterior probability that bentazone is a leacher. The centre of mass under this curve is located at 0·330, the posterior mean for the leaching probability, which we noted above. We might have obtained different results from the lysimeter experiments. Had we seen one leacher out of four ($r = 1$), the posterior distribution would be further to the right because the data begin to cast doubt on bentazone being a non-leacher. The posterior mean for the leaching probability would also be higher, at 0·497. In the event that we see more leachers in four experiments, the posterior distributions are shifted

TABLE 2
Prior Screening and Posterior Probabilities for Bentazone

K_{oc}	$t_{1/2}^{soil}$	*GUS*	*GUS classification*	*Prior leaching probability*	*Prior probability 90% interval*	*Posterior leaching probability*	*Posterior probability 90% interval*
13·3	3	1·37	non-leacher	0·927	0·417–0·978	0·309	0·095–0·607
13·3	21	3·80	leacher	0·999	0·464–1·000	0·333	0·108–0·632
34·0	20	3·21	leacher	0·990	0·458–0·997	0·330	0·106–0·628
175·6	3	0·84	non-leacher	0·0012	0·000–0·537	0·0004	0·000–0·281
175·6	21	2·32	transitional	0·238	0·064–0·683	0·079	0·011–0·368

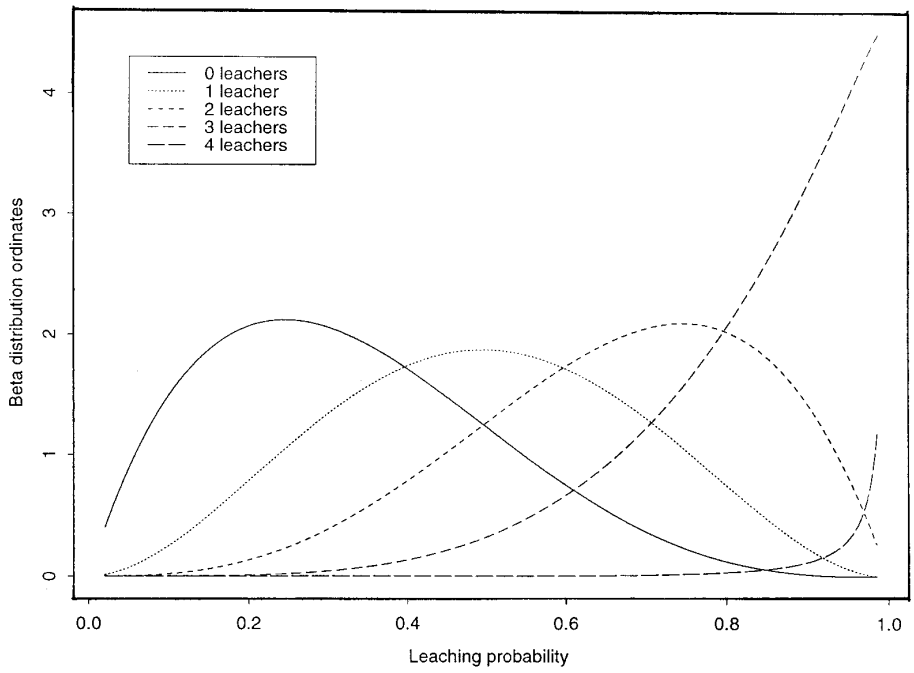

Fig. 3. The five possible posterior distributions for the leaching probability of bentazone based on the number r of the four lysimeter experiments in which it leached. The distributions are drawn assuming that bentazone has an adsorption coefficient of 34 and a soil half-life of 20 days. The actual posterior distribution, corresponding to $r = 0$, is shown by the solid curve; that for $r = 1$ is shown by the dotted curve, etc. Much of the probability area beneath the curve for $r = 4$ leachers is very close to one, and not shown.

further to the right, corresponding to increasing evidence of bentazone being a leacher. The posterior means for the leaching probabilities when we see 2, 3, 4 leachers are 0·663, 0·830, 0·997, respectively.

3.5.3 Aspects of interpretation

This example highlights a number of interpretational problems once the analysis has been carried out. For bentazone, ranges of possible values for the adsorption coefficient and soil half-life are given, rather than single values. The result is a corresponding range of different prior distributions. This would be of little account if the different prior distributions were qualitatively similar, but for bentazone this is not the case: the prescreening offers quite different leaching probabilities at different corners of the $(K_{oc}, t_{1/2}^{soil})$ region. It remains the case that all the posterior leaching probabilities are smaller than 0·34: all four lysimeter experiments lead to the same conclusion of no leaching and these observations tend to dominate the prior information, whatever corner we explore: this is due to our valuation of the prior evidence as worth only $m = 2$ lysimeter experiments.

Although the posterior probability distributions are similar (as in this case they tend to reflect the data rather than whichever prior distribution was used), we should note that there are plausible values of K_{oc} and $t_{1/2}^{soil}$ for which the prior evidence strongly indicates that bentazone leaches, whereas all the lysimeter evidence is to the contrary. Therefore there is a conflict between the data and the prior evidence. There is no alternative here but to re-examine the plausibility of the prior evidence and the information on which it is based, and in parallel to collect more data in the way of further lysimeter experiments and so forth. In general, such conflict between prior knowledge and data is an important consideration in Bayesian methodology and appropriate diagnostic measures have been developed.[34]

The prior evidence is not very helpful here because of uncertainty about the values of K_{oc} and $t_{1/2}^{soil}$. In practice, if we were concerned about the quality of the prior evidence we could diminish its role by, for example, reducing the value of m in (7), although we feel this places an unwarranted trust in the reliability of lysimeter experiments. Alternatively, we can attempt to make the prior knowledge more precise: in this case, by tackling the uncertainty in the values of K_{oc} and $t_{1/2}^{soil}$. This issue, and the general credibility of the reported values of pesticide chemical properties, is important but has received little attention in the literature. In a further paper,[35] we discuss the implications of these uncertainties for prior screening of pesticides.

Finally, note that we are not aiming to establish here exhaustively whether or not bentazone is a leacher, as this would involve the kind of careful and sophisticated analysis that we point to in our discussion below, taking into account a number of different sources of evidence. However, it is worth noting that bentazone was added to the list of compounds for which an analysis was required in the Anglian region in 1993, and was found to exceed 0·1 μg litre^{-1} in three out of 34 groundwater samples: it was not detected at this level in a subsequent study in 1994.[29]

4 DISCUSSION AND FUTURE DEVELOPMENT

This paper shows how modern statistical techniques, in particular Bayesian methods, can be used to address the interpretation of environmental fate and behaviour data for pesticide registration. In doing so we have introduced three principal innovations: the use of logistic regression for pre-screening, the quantification of prior knowledge, and Bayesian methods for combining prior knowledge with the results of lysimeter experiments. We believe that the proposed methodology provides a significant advance on present practice. We now discuss a number of issues concerning this methodology and the potential for further improvement, which we believe to be considerable.

4.1 The role of Bayesian methodology

Bayesian methodology for statistical inference and decision-making has many advantages over classical statistical techniques.[6–11] Its major advantage is that it provides a formal and coherent framework for quantifying expert knowledge and combining it with actual data. In this way, the role of expert judgement, previously used on an *ad hoc* basis, is formalised and placed on a scientific basis. Also, the proper incorporation of expert knowledge strengthens the reliability of inferences or predictions that we might make. This improved precision is of especial importance when the number of actual observations is limited, as is the case for lysimeter experiments. Furthermore, it becomes possible to determine the number of such experiments required to satisfy any given conditions on accuracy.

Is it desirable to use expert knowledge? Expert knowledge is already part of the registration process, as acknowledged by the UK PSD (pers. comm.). Lysimeter experiments and/or computer experiments are believed or disbelieved according to an expert's opinions, and how relevant he/she deems them to be with regard to average soil and climate conditions. This is an important role which should be scientifically and coherently formalised. It is also the case that experts in the field of pesticide pollution usually have useful knowledge on whether or not a pesticide is likely to be polluting, given a set of adsorption or degradation properties and perhaps also the values of other relevant quantities. Again, it seems obvious that such knowledge should not be wasted.

Quantifying expert knowledge usually involves an expert expressing uncertainties about unknowns in the form of prior probability distributions, a process commonly known as elicitation of prior knowledge. There are technical issues involved here which are outside the scope of this paper. For example, we have shown in this paper a crude but useful method of establishing certain aspects of prior knowledge, but we have omitted most of the facets of a more general elicitation process, which would generally entail seeking the advice of a statistical expert. Additionally, there remains the problem of how valuable we believe the expert knowledge to be, and assessing its value is also a matter of expert judgement. Here, we have deemed the prior knowledge to be worth two lysimeter experiments, but this too is an area for which further research should lead to a more refined approach.

The Bayesian methodology presented in this paper generalises to situations involving more than one unknown parameter, for example probability of leaching and degree of toxicity. A further advantage of the Bayesian approach is that it can be used to highlight any differences that arise between the data and the expert knowledge. This carries feedback information which is useful in revealing and pinpointing errors or contradictions in either source of information. Finally, a major advantage of Bayesian methods is that the quantification of expert knowledge and its combination with relevant data lead to probability-based numerical summaries for the uncertainties for the unknowns of interest. Using these summaries, relevant inferences and subsequent decisions can be made comparatively easily. Furthermore, as the numerical descriptions represent a current state of knowledge, they can be combined at a later date with further data. In this way, the Bayesian methodology provides a mechanism for the continual updating of knowledge as new data arrive.

4.2 Comments on pre-screening using logistic regression

Our logistic regression technique results in a good fit and the model seems sensible (Fig. 2). The fit could be improved slightly at the cost of simplicity by using non-linear logistic regression. For example, a curve could be drawn to discriminate between the leachers and the non-leachers in Fig. 2, and we would obtain a different model which would continue to offer probabilities of leaching. However, we believe that there is no good reason at this stage to attempt to improve our current linear logistic regression strategy. Instead, we would do much better to focus on improving the quality of the data used by the logistic regression. As it is, we have only 22 data points from the CDFA survey, and there have been doubts raised about the K_{oc} and $t_{1/2}^{soil}$ measurements for these. Additionally, there are doubts concerning whether or not the CDFA classification into leacher/non-leacher is acceptable. Gustafson[4] addresses some of these concerns in more detail. In particular, the CDFA data relate to Californian soil and climate conditions, and it is not clear how reliable this evidence is for European or other conditions.

We carried out a limited influence analysis by recalculating the model after excluding the three points

worst classified by the model: prometryn, 2,4-D, and lindane. The recalculated model based on 19 observations turned out to give very similar results to the model based on all 22 observations.

In this model we have not attempted to include other covariates, such as hydrolysis half-life. It is likely that this, together with other covariates that we suggest below, could be used to improve predictions. In particular, it is likely that for some compounds the two-covariate model is simply inadequate to describe their environmental behaviour. For example, K_{oc} is not a comparable measure of soil mobility for compounds for which the partitioning behaviour is not well correlated with the organic carbon content of the soil.[14]

4.3 Issues relating to lysimeter experiments

We have used the results of four lysimeter experiments. We are, however, aware of other lysimeter experiments for bentazone (using different application rates or thresholds) which we could not simply incorporate, as there is apparently still no single standard definition of such an experiment in terms of pesticide application rate, leachate threshold, and so forth.[28,32] The BBA threshold (an annual average of 0·1 μg litre^{-1} for lysimeter leachate) is used throughout this paper.[2] In terms of toxicology, a single threshold value for all pesticides, or even a threshold value at all, may not be appropriate, but such levels provide a common reference point. Future developments would need to address such a standard or devise techniques to overcome the difficulties which arise because some experiments are not directly comparable. Also, if experiments are to be performed as part of the registration process, we must pay close attention as to how well a lysimeter experiment relates to actual conditions under the same soil and climate environment, and then to how well any results transfer to different soil and climate types.

The variability of lysimeter experiments is little understood. Indeed. Yon[36] reports that soil is not a homogeneous medium when considered on the scale of physically close replicate lysimeters. It is possible to gain information about variability by replicating the experiments, except that this is expensive. It may be that a number of replications of smaller-volume lysimeter experiments could deliver more precise information more cheaply.

Current lysimeter experiments for a given pesticide are carried out for a small number of different soil types, from which arises the dubious inference of safety or no-safety for a wide range of soil types. Such practices and inferences need validating. One possibility lies in increased use of computer experiments, as well as further statistical modelling using more soil and climate properties.

4.4 Using evidence from computer experiments

Computer simulations can provide an attractive source of further information, especially when genuine data are expensive to observe, as is the case for lysimeter experiments. However, computer experiments are not direct substitutes for real experiments and depend on the model used. It is essential to calibrate the underlying model used in computer experiments with both actual data and expert judgements. At present there seems to be no widely accepted means of assessing the value of computer experiments in relation to other sources of information such as real data and expert knowledge. One simple and reasonable approach would be for an expert to value a computer experiment as worth some fraction of a real experiment, and, if so, this information could be combined readily with the data and expert knowledge in the manner described in this paper. Otherwise, further research is necessary to determine and formalise the role that computer experiments should play. For example, it could be argued that their main use lies in helping experts to identify and focus their expertise for particular combinations of soil and climate type, pesticide properties, and so forth. There is a small but growing literature on statistical analysis of computer experiments,[37] including examples of Bayesian approaches.[38] Elsewhere, some workers are attempting to use geographical information systems in an effort to increase the range of prediction.[5,39]

In time, and using a properly calibrated and monitored Bayesian approach, we believe it likely that computer experiments combined with expert knowledge will become highly valued, and should make the process of understanding pesticide behaviour cheaper, faster, and more reliable.

4.5 The role of decision theory

Bayesian methodology is also attractive from the more general point of view of making decisions under uncertainty. Next to quantifying uncertainty *via* prior distributions (under assumed statistical models if appropriate), making decisions in general involves the use of utility functions, where the overall goal is to make a decision that maximises expected utility,[6] and the Bayesian framework achieves this aim by combining both these ingredients (probability distributions and utility functions) in a coherent way. For practical application, one of the most important aspects of making decisions is to construct a utility function, which summarises the value of each possible outcome, taking into account risks to the environment, economic benefits, and so forth. It is clear that the actual design of lysimeter experiments is an interesting decision problem, not only in determining sample size, but also in considering what other factors might be influential, such as those

noted above. Indeed, the most satisfactory way of assessing the value of experiments is in terms of the amount of information we expect to get from them, in relation to the positive effect the information may have on the expected utility of our optimal decision. This is an interesting area for future applied research. For example, the choices of utility functions can only be considered in detail when related to actual experiments.

With regard to the general practice of lysimeter experiments and inferences for pesticide registration, there are several related problems that can only be solved satisfactorily by using Bayesian decision theory. One aspect which may seem superficially straightforward, but which raises various subtle inferential and analytical issues, concerns the actual decision of whether or not a pesticide is classified as a 'leacher', given various data, computer experiment evidence, and expert knowledge. For example, how do we assess or quantify the risks of classifying a pesticide as a non-leacher for a certain well-defined soil type and other relevant covariates? Part of an approach could be to formulate the pesticide registration process as a decision model, so that probabilities can be used to understand the utility of further sampling. In general, Bayesian decision theory would seem to offer the only avenue capable of providing a justifiable and credible answer.

4.6 Advances in modelling techniques and data quality

We have noted a number of deficiencies in current methodology. We conclude our discussion by commenting on specific areas which need to be addressed in future developments.

Measurements for the covariates used for the prior modelling may not be precise. For example, bentazone has a soil half-life reported to be between three and 21 days, rather than an accepted single value. Such imprecision may best be handled by probabilistic modelling. For example, we could assign a probability distribution over a plausible range of values for bentazone half-life. Such a probability distribution could express a belief that the soil half-life was more likely to be 12 days than three or 21 days, for example. Incorporating such refinements into the methodology is reasonably straightforward but beyond the scope of this paper.

The data used to help us build prior knowledge must be reliable. The Californian data used in this paper are useful, but may not be sufficiently representative for other areas of the world. Additionally, the range of compounds used in Europe, for example, differs from those used in the USA. We recommend that a suitable database be constructed, maintained, and updated. Such a database would take into account, for each pesticide, the range of soil and climate types for which the pesticide has been found to be polluting or not. For example, the HOST classification system[40] could be used to relate to leaching probabilities for pesticides in UK soils. Such a database should aggregate the evidence from existing monitoring programmes; for example. Gustafson[14] notes work in Minnesota, Wisconsin, and Germany. In time, as it becomes more precise and comprehensive, the database should lead to increasingly reliable predictions. Updating such a database involves continual reassessment and recalibration in the light of data, from whatever source.

Aggregation and analysis of results from different experiments on the same pesticide conducted in varied locations is problematical. The necessary methodology, called meta-analysis, is still being developed, but there are several examples of recent applications of Bayesian formulations of meta-analysis in medicine.[41]

Assessing any relationships between the various covariates (adsorption coefficients, solubilities, half-lives, soil structures, climate types, pesticide application rates, and so forth) requires careful attention, preferably using replication to improve precision. This requires quite substantial statistical modelling, but the benefits should considerably outweigh the costs. One benefit will be to improve the reliability of the modelling of prior information, and this may be used to reduce the number of lysimeter experiments required in the registration process. Another benefit will be to improve the reliability of computer experiments.

The outcome of a lysimeter or computer experiment is frequently a volume of leachate collected over a time period, resulting in a simple yes/no definition of leacher according to a given threshold. It should prove possible instead to use leachate breakthrough curves[36] directly, with suitable account being taken of the covariates involved, particularly soil and climate conditions during the experiment. The resulting time series data can be analysed using Bayesian methods.[42,43]

5 CONCLUSIONS

The methods presented here have several advantages over present practice. First, they explicitly utilise expert knowledge that in current practice is at best only used implicitly. Logistic regression, the method used here for quantifying expert knowledge, has many advantages over other pre-screening methods. Secondly, available knowledge and data are combined into a distribution for the probability of leaching that is useful in itself, and that allows for direct comparison with other compounds, including possible reference compounds. Thirdly, the basic methodology is simple and allows information to be added and incorporated at any time.

In formulating our methodology, the shortcomings of the pesticide registration process with regard to environmental fate and behaviour have been highlighted. We conclude that the time is right to re-

formulate the registration process in terms of current statistical modelling and Bayesian technology.

6 ACKNOWLEDGEMENTS

The authors wish to acknowledge the role of Simon P. Cleall (University of Reading) in suggesting to the first-named author the potential of Bayesian methods for analysing environmental fate and behaviour data, and for helpful comments on preliminary versions of this paper. We are grateful to a referee for detailed and constructive comments.

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